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CERAMIC MADE FROM SHS SILICON NITRIDE POWDER

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A ceramic made from a powder of silicon nitride which is obtained by self-propagating high-temperature synthesis is investigated. Yttrium-aluminum garnet and mullite are used as sintering aids. The strength of the material obtained reaches 540 MPa.

When new technical objects are created, as a rule, there also arises a need for new materials, including building materials. The world production of industrial ceramic articles, which are used in a number of sectors of industry as high-performance materials, has grown by one-third in terms of cost from 1999 to 2002 [1]. In the last 20 years new ceramic building materials based on silicon nitride and carbide have been developed and are widely used in developed countries. Investigations have shown that with respect to a complex of properties these materials are greatly superior to other types of ceramic materials. Silicon nitride and carbide and materials based on them have high working temperatures (up to 1700°C), durability, chemical inertness, and high strength and crack-resistance [2], and they have found extensive applications in aerospace, metallurgical, chemical, electronic, and other areas [3]. Materials based on silicon nitride are used in the manufacture of valves for internal combustion engines, turbosupercharger rotors, shut-off fixtures, bearings, cutting tools, seals, and other articles.

At the same time intensive research on further improving the technology for obtaining materials with the objective of optimizing the phase composition and microstructure of monolithic ceramic, developing composite structures, and so on is continuing [4]. A great deal of attention is being devoted to different formation methods: cold and hot slip casting (pressure casting), sintering without applying pressure and under elevated pressure, hot pressing, and hot isostatic pressing. Mass production of articles with complex shapes, high physicochemical properties, isotropic structure, and requiring minimal mechanical working are now on the agenda [5 – 7].

The main problem of obtaining ceramic from silicon nitride is the difficulty of sintering the ceramic [8]. Ultrafine and nano-size silicon nitride powders [9, 10] and sintering activators, often aluminum and yttrium compounds [11, 12], are used to obtain a high-density ceramic.

Our work was devoted to obtaining a dense ceramic from powder of silicon nitride which is obtained by self-propagating high-temperature synthesis (SHS).

Silicon nitride obtained at ISMAN by SHS with α-phase content 95%, specific surface area of the powder 7.4 m^2/g, and average particle size 1 – 2 μm served as the initial material (Fig. 1). The silicon nitride powder contained the following chemical elements besides silicon (%): 38.5 nitrogen, 1.8 oxygen, and 0.05 iron. The integral and differential curves of the grain composition of the powder are presented in Fig. 2.

Silicon nitride obtained by plasma chemical synthesis, containing a mixture of α- and β-phases, with specific surface area of the powder 20 m^2/g and average particle size less than 0.1 μm, was also used as an additive.

Sintering yttrium-aluminum garnet and mullite additives were synthesized separately and added in amounts up to 20%.

Mixing of the powders and additional grinding of the powder obtained by SHS were performed using corundum balls in acetone in teflon drums placed in a planetary mill.

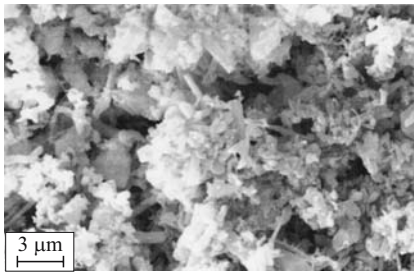

Fig. 1. Electron-microscope photograph of silicon nitride powder obtained by SHS.

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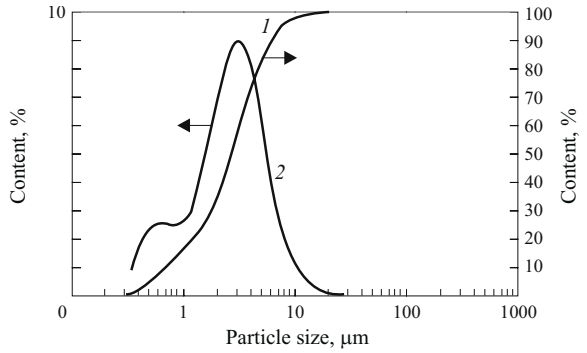

Fig. 2. Integral (*1*) and differential (*2*) curves of the granularity of the initial silicon nitride powder.

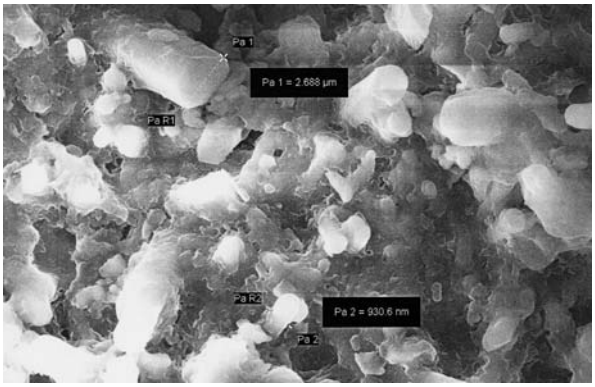

Fig. 3. Structure of ceramic with the addition of 20% mullite and sintered at 1900°C.

The yttrium-aluminum garnet additive was synthesized from a mixture of aluminum and yttrium hydroxides, which was obtained by co-precipitation from water solutions of aluminum and yttrium chlorides in a solution of ammonia. The concentration of the salt solution was 0.95*C*, where *C* is the solubility of the salts at the boiling temperature.

The mixture of hydroxides was filtered, washed with distilled water, and calcined at 1000°C for 2 h. The synthesized yttrium-aluminum garnet had 20 – 80 μm size particles and was subjected to disaggregation in a planetary mill.

A tetraethylsilane sol was used to synthesize the mullite additive. The tetraethylsilane was mixed with a solution of aluminum chloride and precipitated by a solution of ammonia. The concentration of the aluminum chloride solution was 0.5*C*. The mixture obtained was evaporated and calcined at 1000°C for 2 h. The result was a mullite powder with particles less than 1 μm in size.

The samples were pressed into rods in a hydraulic press at pressure 150 MPa. Polyvinyl alcohol was used as a temporary process binder. Next, the samples were placed into a latex jacket and subjected to additional densification at isostatic pressure 220 MPa. After thermogravimetric analysis, a regime was chosen for removing the temporary process binder — calcination at 600°C for 2 h.

The samples were sintered in a mix, consisting of granulated silicon nitride powder, in a nitrogen atmosphere in a high-temperature vacuum furnace in the temperature range 1750 – 1950°C; the holding time at the maximum calcination temperature was 2 h.

The samples obtained were investigated by the methods of petrographic and x-ray phase analysis and by electron microscopy. The density, water absorption, and mechanical strength of the samples were determined.

The density of the samples fluctuated in the range 2.9 – 3.1 g/cm^3, and the open porosity was 1 – 10%. An inverse proportionality was observed between the open porosity and the flexural strength of the material. The flexural strength of the samples obtained from SHS powder did not exceed 450 MPa. The best results were obtained for materials which were sintered at 1900°C and consisted of a 3 : 7 mixture of silicon nitride powders obtained by SHS and plasma-chemical synthesis with 10% additions of mullite and yttrium-aluminum garnet: the density of such samples exceeded 2.95 g/cm^3 and the flexural strength was 520 MPa.

Analysis of the phase composition of the samples showed that the main phase of silicon nitride consisted of isometric silicon nitride crystals 6 – 7 μm (sintering with 10% mullite) and 3 μm in size (with the addition of yttrium-aluminum garnet). In the latter case, 4 – 8 μm prismatic crystals were also present. The additives formed a continuous phase, partially dissolving the silicon nitride grains along their periphery, and the yttrium-aluminum garnet additive formed a phase which crystallized as a solid solution belonging to the cubic system.

The electron-microscope investigations of the samples with the addition of 20% mullite showed that their microstructure consists primarily of a dense glass-like matrix with individual pores 10 – 15 μm in size. The matrix contains prismatic silicon nitride crystals up to 10 μm long and up to 3 μm in diameter (Fig. 3).

In summary, when the content of the additives exceeded 10% the density and strength of the samples decreased as a result of the formation of substantial amounts of secondary phases, including glass phases. The strongest samples were obtained with lesser amounts of additives and were characterized by a uniform distribution of a glass phase with a complex composition and dense intergrowth of silicon nitride crystals.

A ceramic, based on Si_3N_4 powders of different nature, with flexural strength 500 – 540 MPa was obtained as a result of this work.

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